

Title: Two novel biofilters to remove volatile organic compounds emitted by landfill sites

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Abstract

The present work investigates the potential of two experimental field columns (FC-2 and FC-4) to reduce volatile organic compounds (VOC) emissions from municipal solid wastes (MSW) landfill located in Quebec, Canada. The FC-2 and FC-4 were fed by raw biogas coming from the landfill site. The VOC were identified and quantified in emitted biogas and raw biogas. The emitted biogas was collected at the surface of FC-2 and FC-4, and the raw biogas was obtained directly from the well. The main groups of the VOC in the landfill biogas are BTEX (66%), alkanes (19%), cyclics compounds (10%), and halogenated compounds (5%). The concentration of VOC in the landfill raw biogas varies from below the limit of detection (BLD) to 22 ppmv, and that of the emitted biogas varies from BLD to 3.1 ppmv. The result of this study showed that the experimental field columns had a very high potential to reduce the VOC emissions from the investigated landfill. The effectiveness of the VOC emissions removal for the FC-2 and FC-4 was shown to be practically 100% for many compounds. The experimental field columns elimination capacity of VOC emissions is in the range of 0.1 to 4.6 mg.m⁻³.h⁻¹.

Key words: biogas, landfill, volatile organic compounds, biofilters

Introduction

The waste disposal represents a serious problem if biogas emissions are not controlled (Mnif et al., 2012). Broadly, landfills are the most common, simple and economic methods to dispose the wastes (Erses et al., 2008). This method produces biogas emissions into the atmosphere. Decomposition of solid wastes may cause environmental problems, more especially if the biogas emissions are not controlled. Over the last decade much attention has been paid to the biogas emitted by municipal solid waste (MSW) landfills. MSW contains hazardous substances, vehicle maintenance products, mercury-containing waste, certain detergents, personal care products, pharmaceuticals, garden pesticides, batteries and many other industrial wastes (Slack et al., 2005). Landfill biogas (LFG) is produced by microbial degradation of organic matter under anaerobic conditions (Beylot et al., 2012; Chiriac et al., 2007). Biogas is produced in different environments such as: i) in sewage sludge, ii) in bio-waste digesters during anaerobic degradation of organic materials, and iii) in landfills (Rasi et al., 2007). Biogas production depends on the wastes features and age, the available organic biodegradable carbon content and climatic conditions (Desideri et al., 2003; Manna et al., 1999). The LFG content is typically composed of 55–65% v/v methane, 40–45% v/v carbon dioxide, 5–15% v/v nitrogen and 1% v/v for trace compounds (volatile organic compounds (VOC) and odorant compounds) (Albanna et al., 2007; Beylot et al., 2012; Durmusoglu et al., 2010; Johari et al., 2012; Ménard et al., 2012; Rasi et al., 2011; Rasi et al., 2007; Schweigkofler and Niessner, 1999; Shin et al., 2002; Tassi et al., 2009). According to Rasi et al. (2007), amounts of VOC and odorants compounds are low compared to methane. The formation or production of the VOC in landfill can be results from: i) either associated or

competitive secondary reactions or reorganization of the organic matter during the humification, and ii) escaping of VOC present initially in the waste buried in landfill; the biogas is the emission vector (Staley et al., 2006). Many VOC are emitted from landfill (Scheutz et al., 2008). The LFG contained at least these classes of compounds saturated and unsaturated hydrocarbons, acidic hydrocarbons and organic alcohols, aromatic hydrocarbons, halogenated compounds, sulfur compounds such as carbon disulfide and mercaptans and inorganic compounds (Allen et al., 1997). The VOC concentrations depend on the quantity and composition of the solid waste dumped at the site (Mor et al., 2006). The VOC concentration in raw biogas of landfill can range from 0.05 to 1077 mg.m⁻³ (Rasi et al., 2011). Benzene, toluene, ethylbenzene and xylene isomers (known by BTEX compounds) form an important group of VOC found in landfill biogas (Durmusoglu et al., 2010; Hoque et al., 2008).

The VOC emissions are prone to play an essential role in the photochemical processes, which are responsible for air pollution in the atmosphere (Wang et al., 2012; Yu et al., 2012). In the presence of NO_x, the VOC compounds react with OH radicals to form ozone resulting in modifying the chemical reactions in the atmosphere (Hoque et al., 2008). The VOC are harmful to animals and plants, and they contribute to the greenhouse effect (Salvador et al., 2006). Some VOC despite their low concentrations may exert adverse effects on environment and human health (Durmusoglu et al., 2010; Mohan et al., 2006). According to Haque et al. (2012) and Mello et al. (2010), the chronic exposure to BTEX is linked to increased risk of cancer, central nervous system disorders, and damage to blood, liver, and kidneys.

92 Control of VOC emissions from landfill facilities is becoming, therefore, one of the
93 largest environmental challenges facing the municipal waste management to day. Many
94 techniques are commonly employed to reduce the biogas emission from landfill sites,
95 such as installation of daily and final covers, and biogas collection systems. However,
96 none of these systems are 100% efficient (Spokas et al., 2006). Consequently, part of the
97 generated landfill gas and associated VOC can escape to the atmosphere as fugitive
98 emissions. The installation of properly designed biofilters appears to be a promising
99 alternative to reduce fugitive and residual emissions of methane. It was hypothesized in
100 the present study that biofilters could also be effective in reducing VOC emissions. The
101 biotechnology approach is an efficient and potentially alternative for the treatment of
102 VOC compounds (Mello et al., 2010). The most common emission control technology in
103 current use is the installation of biocover on landfill biogas. The microorganisms living in
104 biological systems are the capacity to degrade VOC (Haque et al., 2012). According to
105 Haque et al. (2012) many research remains to be carried out before this technology can be
106 widely implemented. A review of the literature suggests that studies focused on removal
107 VOC in landfill site are rather limited (Allen et al., 1997; Durmusoglu et al., 2010; Haque
108 et al., 2012; Scheutz et al., 2008). To enrich scientific research in this field, two
109 experimental field columns FC-2 and FC- 4 were installed on Saint-Nicephore (Quebec,
110 Canada) landfill site and are fed by biogas coming from the biogas well. This paper
111 documents with field data the reduction of VOC emissions by experimental field columns
112 under the real meteorological conditions. The main objectives of the present work are
113 concerned with the identification and quantification of VOC in raw biogas and emitted
114 biogas collected at the surface of FC-2 and FC-4. The sub- objective is to study the VOC

removal effectiveness and the elimination capacity of FC-2 and FC-4 to reduce VOC contained in LFG under the real conditions. This paper addresses the characterization of the composition of the LFG.

1 Materials and methods

1.1 Description of the field columns

In June 2012, two experimental field columns (FC-2 and FC-4) were constructed in Saint-Nicephore (Quebec, Canada) landfill site (Figure 1).



Fig. 1 Scheme of FC-2 and FC-4

These experimental field columns are 0.90 m (L), 0.90 m (W) and 0.45 m (H). The FC-2 is composed of sand and top soil (Table 1). FC-4 is composed by sand, top soil and compost (Table 1). The FC-2 and FC-4 characteristics are documented in Table 1. The

natural media are the commonly used packing media in the biofilters due to their large surface area, high void fraction, and high microbial population density with sufficient nutrients (Galera et al., 2008; Singh et al., 2010).

The atmospheric conditions in Saint–Nicéphore landfill site were reported in Table 2.

Table 1 Characteristics of the experimental field columns

Field Columns	Substrate	Thickness (m)	Organic matter content (% g _{o.m.} /g _{dry soil})	Density (kg m ⁻³)	Total porosity (%)
FC–2*	Top soil	0.15	5.7	1 209	52
	Sand	0.30	0.8	1 611	41
FC–4*	Mixture of top soil and compost	0.05	9.4	n.d. ^a	n.d. ^{**}
	Top soil	0.10	6.0	1 285	52
	Sand	0.30	0.7	1 526	41

*= Field columns; **=Not determined;

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Table 2 Atmospheric conditions in landfill site

Field column	Sampling dates	Atmospheric pressure (kPa)	48-h. accumulated precipitation (mm)	Temperature (°C)
	07-26-12	99.1	12.6	21.6
FC-2*	08-23-12	100.2	0.0	21.9
and	09-06-12	99.7	3.8	22.1
FC-4*	09-20-12	101.0	23.2	13.3
	10-09-12	101.0	18.4	6.8

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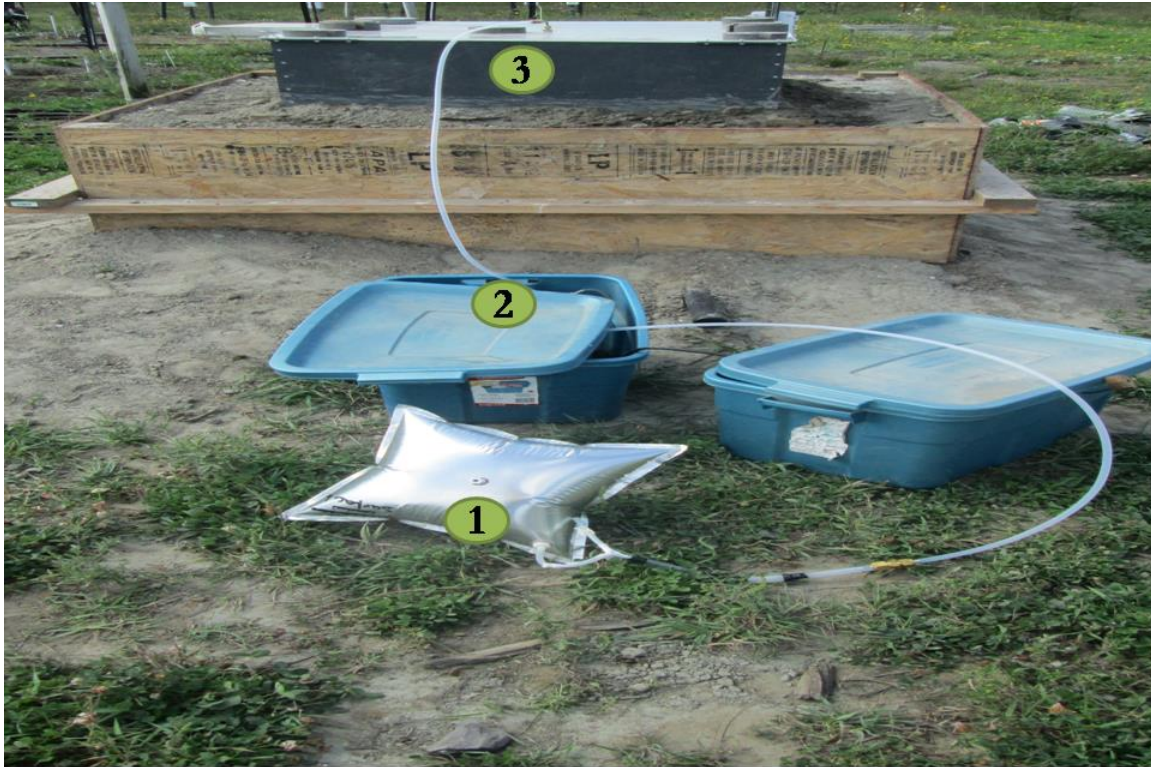
* = Field columns

143 FC-2 and FC-4 were fed by the biogas coming from a capture well. Columns were fed at
 144 different flow rates ranging from 12 to 18 L.h⁻¹ during the sampling period. The sampling
 145 period was took place in Saint-Nicéphore (Quebec, Canada) from 14th August to 30th
 146 September 2012.

147 **1.2 Collection of samples and analyses**

148 **1.2.1 Collection of samples**

149 To collect the emitted biogas from FC-2 and FC-4, a rectangular PVC Flux Chamber
 150 was used (Figure 2).



1= Bag; 2= Peristaltic pump; 3= Flux chamber

Fig. 2 Sampling the emitted biogas from FC-2 and FC-4

A peristaltic pump was used to collect a volume of 10 L of the emitted biogas in the flux chamber. Gas was pumped using a peristaltic pump at a flow rate adjusted to match the methane (CH₄) surface flux (obtained previously to VOC sampling). The CH₄ flux rate was estimated using equation 1 (Barlaz et al., 2004; Scheutz et al., 2008):

$$Flux\ rate = (V/A) * (dC/dt) \quad (1)$$

Where (dC/dt) is the slope of a plot of methane concentration (ppmv) *versus* time (min), V (m³) is the volume and A the area (m²) of the flux chamber.

A volume of 10 L of the raw and emitted biogas was collected directly from the biogas well (Figure 3). Bags for collecting the biogas were made of Tedlar. Tedlar bags have been widely used as a container for the collection of biogas / air samples (Mariné et al.,

2012). Prior to their usage, these bags were ventilated with compressed and filtered ambient air for 12 hours. These bags were then closed with a septum. To ensure accuracy of the results, the bags were checked for background VOC before sampling the biogas. In the field, before connecting the bag to the biogas well, the pipe was flushed for a few minutes. For the emitted biogas at the surface of FC-2 and FC-4, air was allowed to pass through the pipe (Figure 2) for 30 minutes.



Fig. 3 Sampling the biogas from the biogas well

The FC-2 and FC-4 performances were determined with two main parameters: The VOC removal efficiency, RE (%); and the VOC elimination capacity, EC ($\text{g.m}^{-3}.\text{h}^{-1}$); as follows:

$$\text{RE (\%)} = [(C_{\text{RB}} - C_{\text{EB}})/C_{\text{RB}}]*100 \quad (2)$$

$$\text{EC} = (Q/V)*(C_{\text{RB}} - C_{\text{EB}}) \quad (3)$$

Where Q is the biogas flow rate ($\text{m}^3 \cdot \text{h}^{-1}$), V is the field column bed volume (m^3 , including void space), C_{RB} and C_{EB} are the VOC concentration ($\text{g} \cdot \text{m}^{-3}$) in raw biogas and emitted biogas, respectively.

1.2.2 Analyses

A solid-phase micro-extraction (SPME) fibre (Carboxen/PDMS, 85 μm) was used to extract VOC from the collected samples (Cai et al., 2006; Tassi et al., 2009). The fibre was cleaned each time following the same method to treat samples. When using the fibre for the first time, a conditioning step was performed by putting the fibre in the gas chromatography (GC) injector for sixty minutes at a temperature of 300 °C. Then, the SPME fibre was introduced into the sampling bag containing the biogas for 30 min to adsorb VOC. Finally, the fibre was placed in the GC injector for 5 min at a temperature of 290 °C to desorb VOC prior to analysis.

For the detection and quantification of VOC, a GC (G1800A, Hewlett–Packard) equipped with an electron ionization detector [MS] and an HP–5 MS fused–silica column (30 m X 0.25 mm i.d., 0.25 mm film thickness, Hewlett–Packard) were used.

Analysis was conducted in full-scan mode over an m/z range of 50 to 450 amu. Helium was used as carrier gas at a flow rate of 1.20 $\text{mL} \cdot \text{min}^{-1}$. The column was held at 35 °C for 6 min, then the temperature increased to 225 °C at a rate of 20 °C min^{-1} , and it was finally increased to 300 °C at a rate of 30 °C min^{-1} and held for 6 min.

For the quantification of BTEX group, the GC was calibrated by the same method that was used by Wang et al. (1996) and Wu et al. (2004).

Besides BTEX group, four groups (Table 3) of VOC are identified and quantified. The concentration of this groups was expressed in “toluene equivalent” (Chiriac et al., 2011)

because of the high number of the VOC identified and quantified and our analytical possibilities. The BTEX limit of detection (ppbv) was 1.8, 38.1, 4.1 and 24.5 for benzene, toluene, ethylbenzene and xylene, respectively.

2. Results and Discussions

The raw biogas collected at Saint–Nicephore landfill site contained the following group: i) BTEX, ii) alkanes, iii) aromatic compounds (BTEX excluded), iv) cyclic compounds, and v) halogenated compounds. The BTEX compounds are the dominant group in raw biogas (Figure 4). The results obtained in this study are generally comparable with others reported earlier in the literature (Durmusoglu et al., 2010; Scheutz et al., 2008; Tassi et al., 2009).

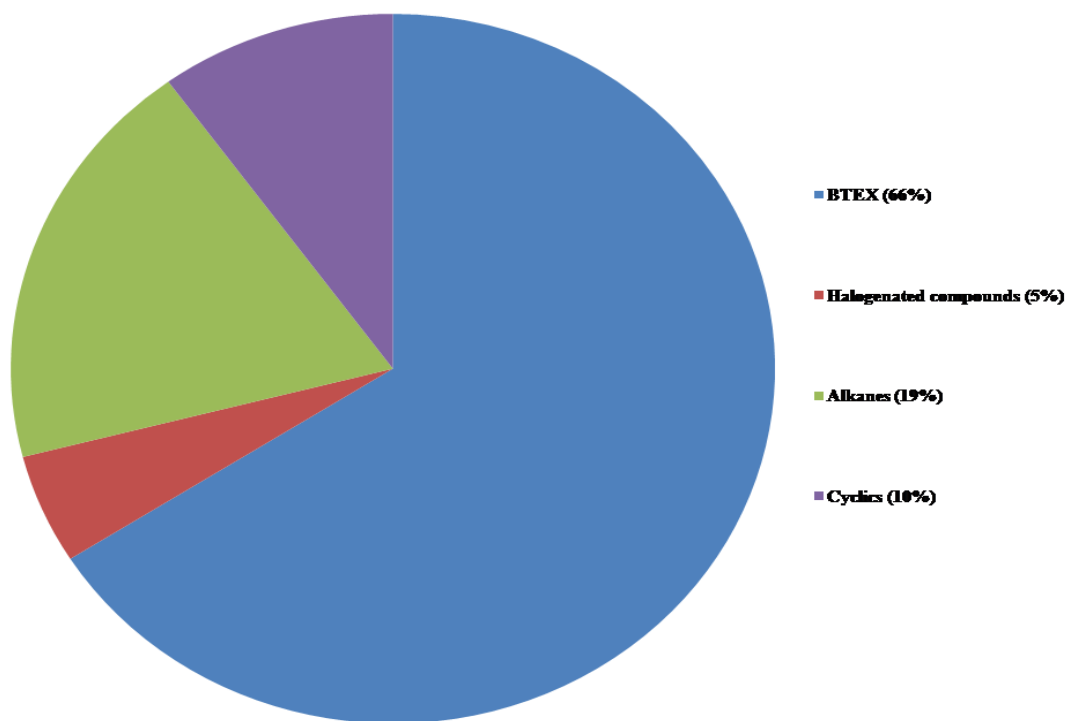


Fig. 4 VOC group identified and quantified in LFG at Saint–Nicéphore landfill site

The VOC concentrations in the raw biogas and the emitted biogas were summarized in the Table 3. In the raw biogas, VOC concentration (ppmv) was in the range of BLD to 22, 0.5 to 3.1, 0.6 to 2.2, and BLD to 1.7 for BTEX, alkanes, cyclic, and halogenated compounds, respectively. Aromatics compounds (BTEX excluded) concentrations are below the limit of detection in the raw biogas.

During the sampling period, 14th August to 30th September 2012, FC-2 and FC-4 are fed with a controlled debit varying from 12 to 18 L.h⁻¹. The concentration of the emitted biogas at the surface of FC-2 and FC-4 was measured for the following chemical groups: BTEX, alkanes, aromatic compounds (BTEX excluded), cyclic compounds, and halogenated compounds. The BTEX concentration varied from BLD to 1.1 and from BLD to 3.1 for biogas collected at the surface of FC-2 and FC-4, respectively. Alkane concentrations (ppmv) in the emitted biogas are in the range of BLD to 0.9 for FC-2 and from BLD to 1.1 for FC-4. For cyclics and halogenated group concentrations are below the limit of detection. For different controlled debits varying from 12 to 18 L.h⁻¹ and during the sampling period, higher concentration value obtained in the present work is for xylene in raw biogas and for toluene in the emitted biogas (Table 3).

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Table 3 VOC concentrations in the raw and in the emitted biogas

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Group	Concentration (ppmv)		
	RB ¹	EB ²	
		FC-2 ³	FC-4 ³
<i>BTEX</i>			
Benzene	BLD ⁴	BLD	BLD
Toluene	11.1 – 19.3	BLD – 1.1	0.5 – 0.7
Ethyl benzene	1.6 – 9.5	0.5 – 0.6	0.6 – 0.7
XyleneΣ (m,p,o)	13.3 – 22	0.6 – 0.8	1.3 – 3.1
<i>Alkane</i>			
Ethane	0.5 – 1.1	BLD	BLD
Propane	0.7 – 1.3	BLD – 0.6	BLD
n-Butane	0.8– 2.1	BLD	BLD
n-Pentane	0.6 – 2.7	BLD – 0.8	BLD – 1.1
Decane	1.7 – 2.9	BLD	BLD
Dodecane	1.5 – 3.1	BLD – 0.9	BLD – 0.6
<i>Aromatics(BTEX excluded)</i>			
1-Methyl 3-ethylbenzene	BLD	BLD	BLD
1,3,4-Trimethylbenzene	BLD	BLD	BLD
1,2,3,4-Tetramethylbenzene	BLD	BLD	BLD
<i>Cyclics</i>			
Methyl cyclohexane	0.7 – 0.9	BLD	BLD
Ethyl cyclohexane	1.0 – 2.2	BLD	BLD
1,2,3-Trimethyl cyclohexane	0.6 – 1.8	BLD	BLD
1,2,4-Trimethyl cyclohexane	0.7 – 0.8	BLD	BLD
<i>Halogenated Compounds</i>			
Dichloro methane	0.8 – 1.7	BLD	BLD
Chloro benzene	BLD	BLD	BLD
Tetrachloro ethylene	0.7 – 0.9	BLD	BLD
1,2-Dichloro ethylene	0.8 – 1.1	BLD	BLD

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¹= Raw Biogas; ²= Emitted Biogas; ³= Field Columns

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⁴= Below the Limit of Detection;

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According to Leson and Winer (1991) and Singh et al. (2006), the RE and EC of biofilters varied with the type of media, airflow rate, inlet loading and the type of VOC to be removed. Only airflow was varied in this study. As shown in Table 4, FC-2 and FC-4 are efficient to reduce the VOC emissions. FC-2 and FC-4 efficiency of the VOC emission are in the range of 54 % to practically 100 %. The VOC removal effectiveness (RE) is in the range of 69 to 100 % and 54 to 100 % for BTEX and alkane. For other groups identified and quantified in LFG, RE is practically 100 % (Table 4).

A number of studies have shown that biofiltration is an efficient biotechnology to reduce VOC emissions (Zarook et al., 1997). High removal efficiency of aromatic hydrocarbons from air can be achieved using biofilters (Zilli et al., 2001). The BTEX removal efficiency of a biofilter composed by coarse sand was 29 % (Haque et al., 2012). In the case of biofilter composed by compost, its removal efficiency to reduce toluene, ethylbenzene and xylene was in the range of 43 to 94 % (Haque et al., 2012). According to Singh et al. (2010) the biofilter removal efficiency for toluene is in the range of 68.2 to 99.9%.

As shown in Table 1, the organic matter content (OM) was determined in FC-2 and FC-4. OM boosts the development of the ubiquitous microorganisms (are not identified in the present work) that can oxidize VOC compounds. The high rates of biofilter efficiencies to reduce VOC emissions obtained can be influenced by a number of factors such as: i) the moisture and the temperature in the biofilter, ii) the soil pH (Lu et al., 2002), and iii) the organic nutrients available in the biofilter soil (Cho et al., 2009; Gaudin et al., 2008; Lu et al., 2002). The organic matter content indicates the presence of nutrients for growth of bacterial (Ait-Benichou et al., 2009; Czepiel et al., 1995). For example, it was reported

that a biofilter composed of natural packing materials like compost demonstrated better performance in the VOC removal compared to soil amendment (Cho et al., 2009). The compost is a desired material for landfill biocovers because it retains moisture for microbial activity (Mostafid et al., 2012). During the sampling period, the atmospheric temperature at the Saint–Nicéphore landfill site varied from 6 to 22 °C. Over the same period, the soil temperature at 10 cm in the FC–2 and FC–4 ranged from 8 to 24 °C. According to Cho et al (Cho et al., 2009), the suitable temperature in biofilters to remove BTEX ranged from 23 to 33 °C.

Table 4 VOC removal effectiveness of FC-2 and FC-4

Group	RE ¹		EC ²	
	FC-2 ³	FC-4 ³	FC-2	FC-4
<i>BTEX</i>				
Benzene	NA ⁴	NA	NA	NA
Toluene	94 – 100	94 – 96	1.3 – 3.4	1.2 – 3.5
Ethyl benzene	69 – 94	62 – 93	0.1 – 1.9	0.1 – 1.9
XyleneΣ (m,p,o)	95 – 96	86 – 90	1.6 – 4.6	1.5 – 4.1
<i>Alkane</i>				
Ethane	100	100	0.1 – 0.2	0.1 – 0.2
Propane	54 – 100	100	~ 0.1	~0.1
n-Butane	100	100	0.1 – 0.4	0.1 – 0.4
n-Pentane	70 – 100	59 – 100	0.1 – 0.3	0.1 – 0.3
Decane	100	100	0.2 – 0.5	0.2 – 0.5
Dodecane	70 – 100	81 – 100	0.2 – 0.4	0.2 – 0.5
<i>Aromatics(BTEX excluded)</i>				
1-Methyl 3-ethylbenzene	NA	NA	NA	NA
1,3,4-Trimethylbenzene	NA	NA	NA	NA
1,2,3,4-Tetramethylbenzene	NA	NA	NA	NA
<i>Cyclics</i>				
Methyl cyclohexane	100	100	0.1 – 0.2	0.1 – 0.2
Ethyl cyclohexane	100	100	0.1 – 0.4	0.1 – 0.4
1,2,3-Trimethyl cyclohexane	100	100	0.1 – 0.3	0.1 – 0.3
1,2,4-Trimethyl cyclohexane	100	100	0.1 – 0.2	0.1 – 0.2
<i>Halogenated Compounds</i>				
Dichloro methane	100	100	0.1 – 0.3	0.1 – 0.3
Chloro benzene	NA	NA	NA	NA
Tetrachloro ethylene	100	100	0.1 – 0.2	0.1 – 0.2
1,2-Dichloro ethylene	100	100	0.1 – 0.2	0.1 – 0.2

281 ¹= VOC removal Efficiency (%); ²= Elimination Capacity (mg.m⁻³.h⁻¹); ³= Field Columns

282 ⁴= Not Applicated

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The FC-2 and FC-4 elimination capacity (EC) was summarized in Table 4. EC ($\text{mg.m}^{-3}.\text{h}^{-1}$) varied from 0.1 to 4.6, 0.1 to 0.5, 0.1 to 0.4, and 0.1 to 0.3 for BTEX, alkane, cyclic compounds and halogenated compounds, respectively. The RE of a biofilter packed with compost and ceramic beads is in the range of 40 % to 95 % and its EC ($\text{g.m}^{-3}.\text{h}^{-1}$) ranging from 3.5 to 128 was observed depending upon the initial loading rates (Rene et al., 2005; Singh et al., 2006).

3. Conclusions

Two field columns are constructed in Saint-Nicéphore landfill site. These field columns are fed by a raw biogas coming directly from the waste mass and under the real meteorological conditions. Saint-Nicéphore LFG contained at least these groups of VOC: i) BTEX, ii) aromatics (BTEX excluded), iii) cyclics, iv), alkanes, and v) halogenated compounds. Among these VOC groups, the BTEX is the dominant. The VOC concentrations ranged from BLD to 22 ppmv. The present work shows that the field columns FC-2 and FC-4 are efficient to reduce VOC emissions. The VOC removal efficiency for FC-2 and FC-4 is in the range of 54 to 100 %. The FC-2 and FC-4 elimination capacity is in the range of 0.1 to 4.6 $\text{mg.m}^{-3}.\text{h}^{-1}$. It can be concluded that the field columns behavior to reduce VOC are practically similar. This work documents with field data the reduction of VOC by experimental field columns under the real conditions. FC-2 and FC-4 represent an interesting technology to reduce VOC emissions from landfill sites.

Globally, it can be concluded that the results obtained during this study shown the potential of field columns to reduce VOC emissions from landfill. However, additional

306 data are required to support the results obtained in the present work. For example, a
307 biologic part should be addressed.

308

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